Remarks on a model of thermal transport in nanofluids

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In a recent letter [1], Kumar et al. outline a model for thermal transport in liquid suspensions of solid nanoparticles, i.e. nanofluids, that purports to explain the anomalous thermal conductivity enhancements observed experimentally. Since this effect is expected to have significant technological consequences and may already be relevant for certain applications [2] this is an important exercise. Unfortunately, the proposal of Kumar et al. falls short on several conceptual counts.

The authors start by considering the case of fixed solid particles and postulate the existence of "two parallel paths of heat flow", one through the suspending liquid and the other through the dispersed solid. This is a very questionable assumption, particularly for the extremely dilute suspensions against which they wish to test their model. By pursuing this argument along an approximate derivation Kumar et al. produce a relation for the effective thermal conductivity of the nanofluid, Eq. 7 of [1], which includes a dependence on the radiuses of the solid particles, r_p, and of the liquid molecules, r_m:

$$k_{eff} = k_m \left[1 + \frac{k_p \epsilon r_m}{k_m (1 - \epsilon) r_p} \right] \qquad (1)$$

,where k_p and k_m are the thermal conductivities of the solid and liquid respectively, and ϵ is the volume fraction of the dispersed solid particles. One problem with the above relation is immediately apparent if we notice that at fixed ϵ, $r_m/r_p \to 0$ implies $k_{eff}/k_m \to 1$, i.e. the solid inclusions have no effect on the thermal conductivity if they are much larger than the liquid molecules. In fact, this situation is rigorously described by theories that treat the liquid as a continuum and where accurate, widely accepted results are available for all ϵ [3]. The dependence of Eq. 1 on k_p/k_m is very different from these results even at $r_m/r_p > 0$. For example, if the solid clusters are much less thermally conducting than the liquid, $k_p/k_m \to 0$, Eq. 1 predicts $k_{eff}/k_m \to 1$, while if they are much more conducting, $k_p/k_m \to \infty$, it yields $k_{eff}/k_m \to \infty$, both independently of ϵ (and r_m/r_p), and both of which can only be deemed incorrect.

A dependence of k_{eff} on the size of the solid inclusions is reasonable, since it can arise for example due to liquid-solid interface thermal resistance [4], even when the liquid can be treated as a continuum. Furthermore, as r_p/r_m decreases the effective volume fraction of the solid particles, e.g. approximated by $\epsilon_{eff} = \epsilon (1 + r_m/r_p)^3$ for $\epsilon \ll 1$, should also perhaps replace ϵ in conductivity estimates, introducing an additional dependence on r_p. However, Eq. 1 captures no such effects and fails some very simple tests, as shown above.

Kumar et al. consider next the effect of the solid particles motion. To this end they invoke kinetic theory and introduce $c\bar{u}_p$ as the "thermal conductivity of the particle", where $\bar{u}_p$ is an average solid particle velocity. Their complete thermal conductivity model is obtained by replacing k_p in Eq. 1 with $c\bar{u}_p$. This is quite problematic since it has the effect of eliminating k_p as a parameter in determining the nanofluid thermal conductivity, and thus makes the previous analysis for fixed solid clusters rather futile. For example, if $\bar{u}_p \to 0$, .e.g. the suspending liquid is frozen, $k_{eff} \to k_m$, i.e. the solid inclusions have no effect on k_{eff} irrespective of k_p and ϵ, when this case should clearly reduce to the fixed particles problem.

In fact, $c\bar{u}_p$ cannot be interpreted at all as the thermal conductivity of a solid particle. By the authors own kinetic theory arguments $k\prime = c\bar{u}_p$ is the thermal conductivity of a dilute gas of particles that possess internal energy [5]. In principle, a quantity like $k\prime$ could provide an estimate for direct, Brownian motion transport of heat by the solid clusters, but it should not supplant k_p, the thermal conductivity of the solid. Unfortunately, the $k\prime$ calculated in [1] is inadequate even for this purpose. The authors estimate of the "constant" $c \propto nlc_v$ (which incidentally is not dimensionless as conveyed in the paper), with n - number density of the solid particles, l - their "mean-free path" and c_v - heat capacity of a solid particle, assumes $l \propto 1/nd_p^2$, $d_p = 2r_p$, which only takes into account the rare collisions between the solid particles and ignores the effect of the liquid. A more reasonable estimate is $kl \simeq nDc_v$, with $D = k_B T/3\pi\eta d_p$ - Stokes-Einstein relation. This yields for $d_p = 10nm$ gold particles in water at normal conditions and $\epsilon = 1\%$, $kl/k_m \simeq 10^{-6}$. Therefore, the direct Brownian motion contribution to thermal transport can be safely ignored for nanofluids, as also pointed out in [6]. Finally, the theoretical values plotted by the authors in their Fig. 4 are off by orders of magnitude from their own formula Eq. 10, purportedly used to calculate them.

In conclusion, the model introduced by Kumar et al. has serious deficiencies and therefore the question of thermal transport in nanofluids remains an open matter. This work was performed under the auspices of the U. S. Department of Energy by University of California Lawrence Livermore National Laboratory under Contract No. W-7405-Eng-48.

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